Electronic Supporting Information

All-polymer indoor photovoltaics with high opencircuit voltage

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Experimental Section

Materials. CD1 (M_n = 35.5 kDa, PDI = 2.75) and PBN-10 (M_n = 52.3 kDa, PDI = 1.66) were synthesized in the laboratory according previously reported methods.^{1,2} ITIC was purchased from Solarmer Materials, Inc.

Characterization. The UV-vis absorption spectra of CD1, PBN-10 and ITIC thin films were measured with a Perkin-Elmer Lambda 35 UV-vis spectrometer. All the film samples were obtained by spin-coating their chloroform solutions. The surface morphologies of CD1:PBN-10 and CD1:ITIC blend films were characterized using atomic force microscopy on a SPA 300HV with a SPI 3800N controller (Seiko Instruments, Inc., Japan) in tapping mode. A silicon micro cantilever (spring constant 2 N m⁻¹ and resonance frequency ca. 300 kHz, Olympus Co., Japan) with an etched conical tip was used for the scan. The transmission electron microscopy (TEM) measurement was performed on a JEOL JEM-1400 transmission electron microscope operating at 120 kV. The two-dimensional grazing incidence wide angle X-ray scattering (2D-GIWAXS) measurements were performed at beamline 7.3.3 at the Advanced Light Source.³ CD1:PBN-10 blend films were prepared by spin-coating their active layer solutions as those used in devices on Si substrate. The 10 keV X-ray beam was incident at a grazing angle of 0.12°-0.16°, which maximized the scattering intensity from the samples. The scattered X-rays were detected using a Dectris Pilatus 1M photon counting detector. The thickness of films was measured with a XP-plus Stylus Profilometer.

Photovoltaic device fabrication and measurement. ITO glass substrates were cleaned by sequential ultrasonication in detergent, deionized water, acetone, and isopropyl alcohol, followed by drying at 120 °C for 30 min and treating with UV-ozone for 25 min. Then the PEDOT: PSS solution (Clevios VP Al 4083 from H. C. Starck Inc.) was spin-coated on the ITO glass substrates at 5000 rpm for 40 s to give a thickness of 40 nm, and baked at 120 °C for 30 min. The PEDOT:PSS substrates were transferred to a nitrogen-filled glove box. CD1 and PBN-10 with a weight ratio of 1:1 were dissolved together in chloroform with a total concentration of 6 mg mL⁻¹, and the solution was stirred at room temperature for 12 h to ensure the complete dissolution. Before spin-coating, 2% by volume of 1-chloronaphthalene was added to the solution and left to stir for 30 min. CD1 and ITIC with a weight ratio of 1:1 were dissolved together in chloroform with a total concentration of 12 mg mL⁻¹, and the solution was stirred at room temperature for 12 h to ensure the complete dissolution. The active layer solutions with different preparing methods were spin-coated on the PEDOT:PSS substrates at spin-rates of 800-1000 rpm to give an active layer thickness of ca. 90 nm. CD1:PBN-10 and CD1:ITIC were annealed at 80 and 100 °C, respectively. At a pressure of 2×10^{-4} Pa, Ca (20 nm) and Al (100 nm) were sequentially deposited on the top of the active layer to complete the devices. The active area of the devices was 8 mm². A XES-40S2-CE class solar simulator (Japan, SAN-EI Electric Co., Ltd.) was used to provide the AM 1.5G simulated solar light illumination. The light intensity was calibrated to be 100 mW cm⁻² using a certified standard monocrystalline silicon (Si) solar cell (SRC-2020, Enli Technology Co., Ltd.). The lux levels of the FL and LED

lamps were measured using a TES-1332A luxmeter (TES Inc., Taiwan) and the intensity calibration was carried out following the previous reports.⁴ The J-V plots of the device were measured with a voltage step of 0.01 V and delay time of 20 ms at 25 °C in a glove box filled with nitrogen (oxygen and water contents are smaller than 0.1 ppm) on a Keithley 2400 source meter. A solar cell spectral response measurement system QE-R3011 (Enli Technology Co., Ltd.) was used to characterize the EQE spectrum under the short-circuit condition. The chopping frequency is 165 Hz.

Hole- and electron-only devices fabrication and mobility measurements. The hole and electron mobilities were measured by SCLC method. The hole-only device structure is ITO/PEDOT:PSS (40 nm)/active layer/MoO₃ (10 nm)/Al (100 nm) and the electron-only device structure is ITO/ZnO (30 nm)/active layer/Ca (20 nm) /Al (100 nm), respectively. J-V plots in the range of 0–10 V were measured using a Keithley 2400 source meter, and the mobility was obtained by fitting the J-V plot near quadratic region according to the modified Mott-Gurney equation:⁵

$$J = \frac{9}{8} \varepsilon_{\rm r} \varepsilon_0 \mu \frac{V^2}{L^3} \exp \mathbb{2} 0.89 \beta \frac{\sqrt{V}}{\sqrt{L}}$$

Where J is the current density, ε_0 is permittivity of free space, ε_r is the relative permittivity (assumed to be 3), μ is the zero-field mobility, V is the potential across the device ($V = V_{applied} - V_{bi} - V_{series}$), L is the thickness of active layer, and β is the fieldactivation factor. The series and contact resistance of the device (10–20 Ω) were measured using blank device of ITO/PEDOT:PSS/MoO₃/Al or ITO/ZnO/Ca/Al.



Fig. S1 Space-charge-limited J-V plots for the hole-only device of (a) CD1 film, and for the electron-only devices of (b) PBN-10 films.



Fig. S2 J-V plots of the OSCs based on CD1:PBN-10 blend film under (a) FL and (b) LED with different light intensities.

Table S1 Photovoltaic parameters of the OSCs based on CD1:PBN-10 blend film

under FL and LED with different light intensities.

Light source	Light intensity (lux)	V _{oc} (V)	J _{SC} (mA cm ⁻²)	FF (%)	PCE (%)	P _{out} (mw cm ⁻²)	P _{in} (mw cm ⁻²)
	200	1.07	0.024	63.0	23.4	0.016	0.069
	500	1.11	0.058	65.5	24.3	0.042	0.173
	1000	1.14	0.120	66.2	26.2	0.091	0.345
ΓL	2000	1.16	0.244	66.7	27.4	0.189	0.690
	4000	1.18	0.452	66.1	25.5	0.352	1.380
	6000	1.19	0.657	65.8	24.9	0.514	2.07
	200	1.07	0.022	62.3	20.4	0.015	0.072
	500	1.11	0.054	64.4	21.4	0.039	0.180
	1000	1.14	0.105	65.4	21.7	0.078	0.360
LED	2000	1.16	0.216	66.4	23.1	0.166	0.719
	4000	1.18	0.426	65.4	22.5	0.323	1.438
	6000	1.19	0.623	65.1	22.4	0.484	2.157



Fig. S3 *J*–*V* plots of the OSCs based on CD1:ITIC blend film under (a) FL and (b) LED with different light intensities.

Table S2 Photovoltaic parameters of the OSCs based on CD1:ITIC blend film under

FL and LED with different light intensities.

Light source	Light intensity (lux)	V _{oc} (V)	J _{SC} (mA cm ⁻²)	FF (%)	PCE (%)	P _{out} (mw cm ⁻²)	P _{in} (mw cm ⁻²)
	200	0.73	0.024	65.7	16.7	0.012	0.069
	500	0.75	0.059	67.0	17.1	0.030	0.173
-	1000	0.78	0.116	68.1	17.9	0.062	0.345
FL	2000	0.80	0.234	68.4	18.6	0.128	0.690
	4000	0.82	0.482	67.9	19.4	0.268	1.380
	6000	0.84	0.689	67.6	18.9	0.391	2.070
	200	0.73	0.021	66.0	14.1	0.010	0.072
	500	0.75	0.053	67.2	14.8	0.027	0.180
	1000	0.77	0.107	67.5	15.4	0.056	0.360
LED	2000	0.80	0.207	68.5	15.8	0.119	0.719
	4000	0.81	0.418	69.7	16.4	0.236	1.438
	6000	0.83	0.613	68.3	16.1	0.348	2.157



Fig. S4 Space-charge-limited J-V plots for the (a) hole-only and (b) electron-only devices of CD1:PBN-10 blend film.



Fig. S5 (a) $J_{ph}-V_{eff}$ plot and (b) light intensity dependence of J_{SC} (solid lines are fitted result) of the photovoltaic devices based on CD1:PBN-10 blend film.



Fig. S6 1D intensity profiles of the GIWAXS patterns for the CD1:PBN-10 and CD1:ITIC blend films in out-of-plane and in-plane directions.



Fig. S7 AFM (a) height and (b) phase, and (c) TEM images of CD1:ITIC blend film.

Photovoltaic cells	E_{g} (eV)	$V_{\rm OC}$ (V)	PCE (%)	References
a-Si	1.63	0.88	20	[6]
μc-Si	1.12	0.47	12	[6]
GaAs	1.43	0.66	19.4	[7]
InGaP	1.86	0.98	30.3	[8]
Perovskite	1.55	0.87	35.2	[9]
DSSC	1.72	0.88	31.8	[10]
Organic	1.82	0.79	28.1	[11]
All-polymer	1.93	1.16	27.4	This work

Table S3 The IPV efficiency and V_{OC} distributions of different types of photovoltaic cells.

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